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ORIGINAL ARTICLE

Evaluation of concrete self-healing by encapsulated sodium metasilicate in perlite and expanded clay

Avaliação de concreto autocicatrizante através do encapsulamento de metasilicato de sódio em perlita e argila expandidas

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Abstract: Investigating the behavior of self-healing cementitious composites is necessary to know alternatives that can be applied in structures increasing their life service. Therefore, this study evaluated concrete self-he aling from the use of expanded perlite (EP) and expanded clay (EC) capsules impregnated with a sodium metasilicate solution. These materials were used to substitute natural aggregates in concrete in proportions of 0 wt·%, 15 wt·% and 30 wt·% which were cured in humid or submerged environments. Cracking was induced with a flexural test and a closing with cicatrization product. was evaluated and measured visually with a software. Capillary absorption tests indicated a reduction in the porosity of samples which incorporated self-healing materials, considering it as an important property related to durability. Samples with EP achieved 100% self-healing with 15% substitution. Crack filling was achieved in cracks up to 0.43 mm wide. Samples with EC achieved 50% crack recovery under humid curing and 90% under submerged curing. It was concluded that incorporating the sodium silicate allowed improvements to fissure sealing and it is an alternative to produce self-healing concrete in Brazil. EP was more effective than EC as encapsulating material. Despite that, the EP did not impact the compressive strength due to its small size and better packing of the mixture, Also, EP presented higher healing percentage when comparing with samples containing EC.

Keywords: concrete, self-healing, expanded perlite, sodium meta-silicate.

Resumo: Investigar o comportamento de compósitos cimentícios autocicatrizantes é necessário para conhecer alternativas que possam ser aplicadas em estruturas aumentando a sua vida útil. Portanto, este estudo avaliou a cicatrização do concreto a partir do uso de cápsulas de perlita expandida (PE) e argila expandida (AE) impregnadas com solução de metassilicato de sódio. Esses materiais foram utilizados em substituição aos agregados naturais no concreto nas proporções de 0%, 15% e 30% em relação à massa. Os exemplares foram curados em ambientes úmidos ou submersos. A fissura foi induzida pelo teste de flexão e a cicatrização foi avaliada visualmente e medida por meio de software. Testes de absorção capilar indicaram redução da porosidade das amostras que incorporaram materiais autocurantes, sendo esta uma importante propriedade relacionada à durabilidade do conjunto. As amostras com PE alcançaram 100% de cicatrização com 15% de substituição. O preenchimento da fissura foi obtido em fissuras de até 0,43 mm de largura. Amostras com AE alcançaram 50%

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Data Availability: The data that support the findings of this study are available from the corresponding author, FP, upon reasonable request.

de recuperação de fissura sob cura úmida e 90% sob cura submersa. Conclui-se que incorporar a solução de metassilicato de sódio proporcionou melhorias consideráveis na selagem de fissuras e esta solução torna-se uma alternativa eficiente para concretos autocicatrizantes a serem produzidos no Brasil. A incorporação de PE foi mais eficaz do que o AE como material encapsulante. Além disso, o uso de PE não impactou na resistência à compressão em decorrência de sua reduzida dimensão de partícula e empacotamento. Ainda, a EP apresentou maior percentual de cicatrização comparando-se com as amostras contendo AE.

Palavras-chave: concreto, autocicatrização, perlita expandida, metassilicato de sódio.

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1 INTRODUCTION

Concrete is widely used worldwide due to its well-known properties. Hardened concrete has its durability linked to its main property of mechanical strength [1]. The U.S. Geological Survey [2] estimated a world production of cement of 4.1 billion metric tonne in 2019. In 2021, world cement production achieved 4.4 billion metric tonne [3]. Cement emissions amount to 1 t CO_2 / t of cement produced [4]. In 2019, worldwide CO_2 emissions were 36.8 ± 1.8 Gt [5], of which an estimated 11.1 ± 0.5% can be attributed to cement production, and thus, is required to increase the lifespan of construction elements that use cement.

Structural concrete is highly susceptible to the formation of cracks. This is a result of low resistance to tension and other mechanisms such as contraction from drying, thermal cycling and shifting, overloading, reinforcement corrosion etc [6]. The reduced durability due to cracks, repair and recovery cost of structures [7] and environmental impact of cement production are driving motivations to the development of self-healing types of concrete, which operates [8]–[11] under the same principle as a human body: injuries cause the release of healing agents which induce regeneration [12]. As noted succinctly by Van Tittelboom et al. [13], there are 3 types of concrete self-healing processes or mechanisms: autogenous, autonomous and vascular.

Autogenous self-healing is a natural phenomenon of concrete, related to delayed cement hydration and/or pozzolan addition. Autonomous healing is obtained from the addition of micro-capsules impregnated with healing agents. When exposed to the environment, these agents induce healing and they are not commonly used in concrete. Lastly, vascular healing also makes use of healing agents but in hollow tubes that connect the external environment to the inside of the structure. Studies have demonstrated that autogenous self-healing materials were limited to small cracks and were thus restricted and less reliable since they could not repair multiple cracking phenomena [13], [14]. As an example, the range of values in which cicatrization happens is between 50μm and 200μm [13].

Autonomous self-healing can be classified as bacterial precipitation or healing induced by chemical agents [12]. Among the chemical healing agents, there is a prevalence in the use of sodium metasilicate (Na_2SiO_3) [6], [7], [15]. This material reacts with calcium hydroxide ($Ca(OH)_2$) inside concrete to produce calcium silicate hydrate (C-S-H) gel which binds to the concrete, partially filling the crack and recovering the strength of the material [16].

For an effective healing process, capsules must remain intact after fresh cement has been mixed, resisting impact from the concrete mixer and other aggregates. It has been noted that spherical capsules were more resistant and influenced the least the workability of concrete under mixing [17]. Capsule wall thickness was also an important factor since thinner walls could result in premature rupture while thicker walls might impede the agent release [12]. Sisomphon et al. [18] made use of expanded clay (EC) capsules with a protective coating of cement. The cement coating was completely cured prior to mixing and the final result was a much improved concrete quality with self-healing properties. Zhang et al. [6] made use of capsules between 2 mm and 4 mm in diameter impregnated with *Bacillus cohnii* and coated in a geopolymer produced from metakaolin and sodium metasilicate.

Aggregates are used as capsules for healing agents but while more capsules increase the probability that a crack might rupture them, too many capsules could increase the cost and compromise the strength of the resulting material [19]. The width of the crack healed varied with respect to the encapsulating method. In the case of spherical capsules, their diameter must be large enough to store and release the healing agent into the crack. This recommended diameter varied between 5 μm and 5 mm. If the crack were too wide, the capsule healing agent could be rapidly expanded [13].

Another factor in the encapsulated healing agent was the local pressure during the manufacturing process. Sisomphon et al. [18] used sodium monofluorophosphate (Na_2PO_3F) in EC capsules 4 mm in diameter. In this case, the absorption rate under natural pressure conditions was lower when compared to absorption in a vacuum. Results showed that the use of a vacuum chamber was a necessity to allow higher agent absorption. Sisomphon et al. [18] also demonstrated that sodium monofluorophosphate healed the samples and significantly improved the quality of concrete in the carbonation region.

Alghamri et al. [20] used light aggregate capsules impregnated with sodium metasilicate. Cracking tests demonstrated an 80% recovery in load-bearing capacity when compared to a control sample. Capillary water absorption was also improved, indicating a reduction in cracking and an expected longer durability of the material.

Pelletier et al. [16] used polyurethane capsules also impregnated with sodium metasilicate. This light aggregate was used as 2% substitution in volume of concrete. Healed samples had strengths up to 10% higher than reference samples. Tan et al. [7] also used polymer capsules with a silica solution at its core. The resulting material had an increase of 7.5% in strength and flexural resistance.

The viscosity of the healing agent is an essential parameter to prevent its absorption into the concrete matrix since it could allow it to flow from inside the capsules. Dry, apud Van Tittelboom et al. [21] concluded that the healing agent viscosity must remain between 100 mPa·s and 500 mPa·s but other studies have noted that lower viscosity agents were able to fill both micro and macro-cracks. Methyl methacrylate was an example of an inappropriate low viscosity agent: cracks might be left open since its curing time was of approximately 30 mins, which was sufficient time for concrete to absorb or leak out part of it. Van Tittlelboom and De Belie [22] increased the viscosity of the healing agent with the addition of poly(methyl methacrylate) as a thickening agent to hold the chemical inside the capsule. Viscosity was also associated with the SiO_2/Na_2O molar ratio (also known as the silica modulus). Higher silica modulus indicated a more viscous agent and, for most healing agents, varied between 1.60 to 3.75 [23].

The most used encapsulating aggregate is expanded clay (EC) since it is readily available and of low cost but with low mechanical resistance. Another light aggregate is expanded perlite (EP) which is also readily available and less expensive than other aggregates such as vermiculite, pumice etc [24]. The use of EC as encapsulating material for bacterial spores was tested by Jonkers [25]. It was determined that cracks of up to 0.46 mm were healed after 100 days curing in water immersion. While viable, this technique also increased the porosity of concrete and reduced its resistance. In the case of 50 wt·% aggregate substitution with EC, samples presented a 50% reduction in resistance after 28 days of curing, which was not recommended for structural applications [12], [24].

Further studies were conducted to improve autonomous self-healing capacity. Zhang et al. [6] compared the use of EC and EP as capsules for Bacillus cohnii bacteria. After 28 days curing, EP and EC were able to recover cracks up to 0.79 mm and 0.45 mm wide, respectively.

Sisomphon et al. [18] demonstrated that self-healing with microencapsulated sodium monofluorophosphate delayed water penetration and increased concrete strength. In more detail, the healing agent increased strength for concrete subjected to a high stress environment which included freezing/unfreezing cycles and exposure to salinity. Sodium, phosphorus and fluorine were also detected in the concrete matrix which proved the successful release of the healing agent from the capsules.

Even knowing that several studies have already been performed in this area, the present study compares the use of sodium silicate when using percentages of 10, 20 e 30% of expanded perlite and expanded clay. The aim is to evaluate procedures viable to be applied on a large scale for real concrete procedures. The present study used EC and EP as transport media for the healing agent. Perlite is a volcanic silicate glass which can expand as much as 20x in volume when rapidly heated, leaving several internal void spaces [19]. Expanded clay has been widely studied and is already used in structural concrete [26]. Nevertheless, EC has enough void spaces to be used in self-healing concrete [27]. The chosen healing agent was sodium metasilicate encapsulated in either EC or EP and an outer protective coating. Evaluation techniques included examination of the microstructure of both aggregates and varying the type of curing: samples were immersed in water while others were exposed to a humid environment. Mechanical and physical tests were conducted and the formation of healing products verified with scanning electronic microscopy (SEM).

2 MATERIALS AND METHODS

The methodology consisted of the production of 5 compositions: a reference concrete and concretes with 15 wt·% and 30% wt·% substitution of sand with EP and EC aggregates impregnated with sodium metasilicate.

2.1 Materials and composition

The Portland cement used was of type III as defined in ASTM C150 [28]. Fine aggregate was locally sourced river quartz sand. Sand bulk density was 1,411 kg/m^3 while specific mass was 2,595 kg/m^3, as determined from ASTM C29 [29] and ASTM C128 [30] procedures, respectively. The granulometric distribution of the sand was determined following ASTM C33 [31] procedures and is shown in Figure 1. The resulting fineness modulus was 3.07 and the maximum dimension was 2.36 mm.

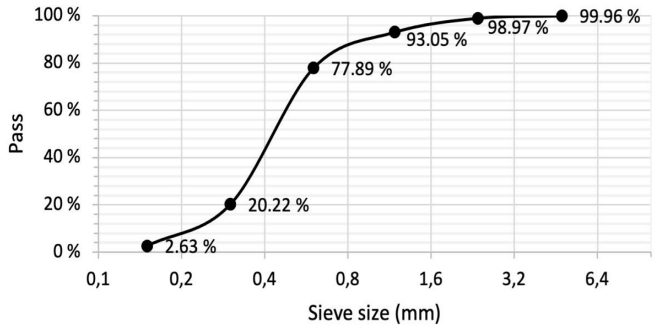

Figure 1 - Granulometric distribution of fine aggregate

The EP used had granulometry between 1.2 mm and 4.8 mm with bulk density of 110.1 kg/m^3. The EP aggregate showed a mass of 94.36 kg/m^3 in the natural state and after being impregnated with chemical solution the value was 475.50 kg/m^3. No further granulometric distributions were examined since fine aggregate substitution was based on the % diameter of sand removed. The EP used was of two types: one with granulometry between 1.2 mm and 4.75 mm and the other between 2.4 mm and 12.5 mm. Substitute aggregate microstructure and chemical composition were evaluated with a Zeiss-brand scanning electronic microscope with energy-dispersive spectroscopy (SEM/EDS). Samples were pre-dried and coated in gold prior to testing and the results are shown in Figure 2.

The SEM/EDS results of Figure 2 show that EP had a more porous surface more likely to absorb the healing agent efficiently. In contrast, EC had a more compact and dense structure than the samples used by Ahmad et al. [32] and Bogas et al. [33] in addition to being fundamentally different from EP. Chemical compositions obtained from EDS in the same regions where the images were taken are shown in Table 1.

Table 1 shows that both substitute aggregates were composed mainly of oxides and silicates, as identified by the O and Si contents, respectively. Both also contained sodium but in less than 3 wt·% which makes it improbable that they contained naturally formed sodium metasilicate. This confirmed the need to impregnate the substitute aggregates with a chemical solution to promote C-S-H formation.

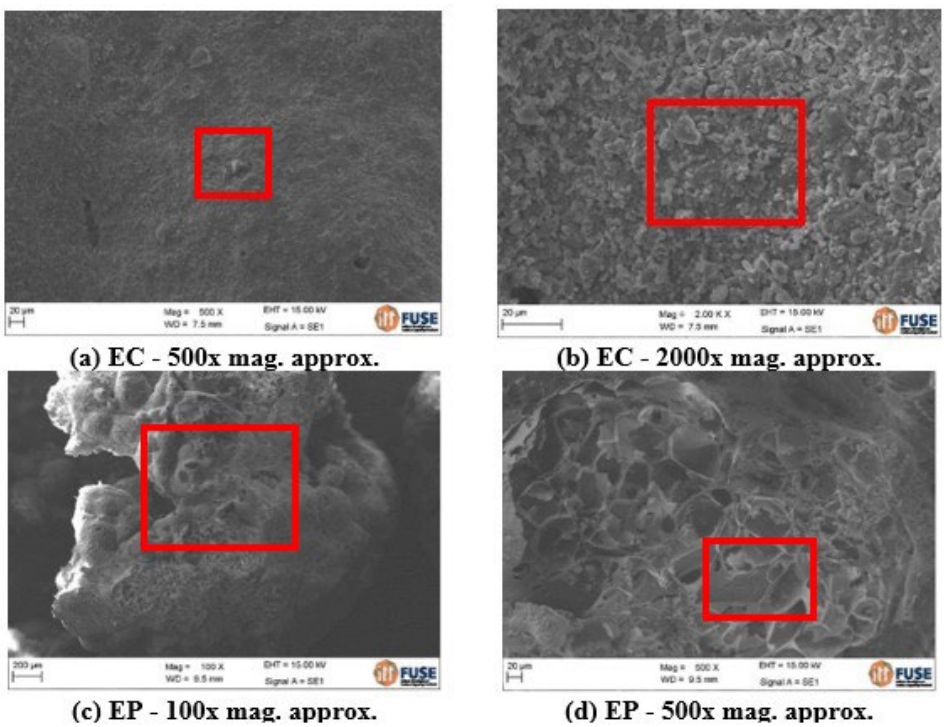

Figure 2 - SEM/EDS images for EC (a,b) and EP (c,d)

Table 1 - EDS semiquantitative chemical analysis of substitute aggregates

Element	EC (%)		EP (%)	
	Weight	Atomic	Weight	Atomic
O	42.80	58.92	43.05	57.65
Na	0.43	0.41	2.27	2.12
Mg	1.78	1.61	0.00	0.00
Al	10.23	8.35	7.18	5.70
Si	30.87	24.22	40.70	31.04
K	4.48	2.52	4.76	2.61
Ca	1.16	0.64	0.62	0.33
Ti	1.15	0.53	0.00	0.00
Fe	7.10	2.80	1.41	0.54
Total	100.00		100.00	

The EC was also applied in replacement of natural fine aggregate. In accordance with its manufacturer, the EC is divided into small and large, considering the specific mass of 850 kg/m^3 and 600 kg/m^3, respectively. Coarse aggregate used was basaltic gravel of granulometry between 4.8 mm and 9.5 mm. The same characterization procedures applied to the quartz river sand fine aggregate were applied to the gravel. The bulk density and specific mass were determined as 1,641 kg/m^3 and 2,480 kg/m^3, respectively. The granulometric distribution of the coarse aggregate is shown in Figure 3. The fineness modulus was determined as 2.57 and the maximum characteristic dimension was 9.5 mm.

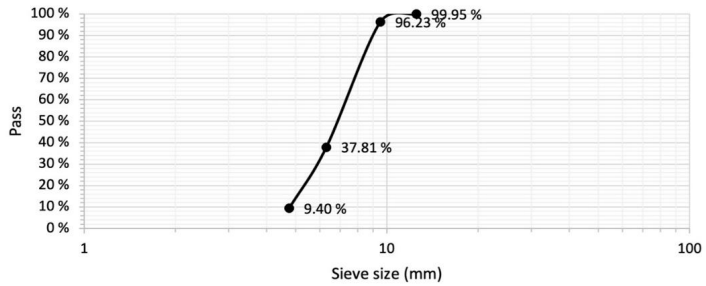

Figure 3 - Granulometric distribution of coarse aggregate (basaltic gravel)

The healing agent used in this study was the same sodium metasilicate (Na_2SiO_3) used in other reference studies [6], [7], [22]. The sodium silicate is from Simoquímica brand, with original concentration of 50%, diluted in water also by 50% The molar ratio SiO_2/Na_2O is 3.27. The healing agent was used in liquid form with 50 wt·% dilution in deionized water. The vacuum impregnation technique of Sisomphon et al. [18] was used to impregnate the healing agent in the substitute aggregates. The original healing agent had viscosity of 496 mPa·s and a SiO_2/Na_2O ratio of 3.27. Other properties are shown in Table 2, through data collected from the manufacturer, and were in accordance with reference work [21].

Table 2 - Sodium metasilicate properties

Property	This work	Reference value
Appearance	Viscous liquid	
Baumé scale (ºBé)	38.50 - 41.50	39.92
Specific mass (g/cm^3)	1.37 - 1.42	1.38
Viscosity (mPa·s)	250 - 500	496
Sodium oxide (%)	8.0 - 9.20	8.43
Silicon dioxide (%)	26.00 - 29.50	27.65
SiO_2/Na_2O molar ratio	3.00 - 3.35	3.27
pH	10.5-12.2	11.2

2.2 Healing agent encapsulation

Healing agent encapsulation consisted of 4 steps:

Step 1: sodium metasilicate was separated according to granulometric size and diluted in 50-wt·% deionized water. Substitute aggregate capsules were submersed in this solution for 6 h to induce pre-saturation;

Step 2: the substitute aggregate capsules and sodium metasilicate solution were exposed to a vacuum for 2 h in a glass desiccator;

Step 3: substitute aggregate capsules were weighted and coated in Portland cement for mechanical protection as described in Sisomphon et al. [18]. The before and after visual aspects are shown in Figure 4ab for EC and Figure 4cd for EP;

Step 4: substitute aggregate capsules were cured in a humidity chamber at 100% humidity for 5 days for later molding.

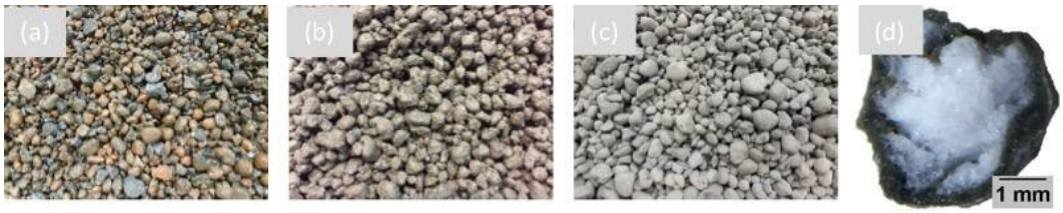

Figure 4 - Before and after cement coating application on substitute aggregate capsules for EC (a, b) and EP (c, d)

2.3 Mixing, Curing and Molding

The samples and mixing ratios used are shown in Table 3. Sand substitution was performed by removing the target wt·% and replacing it with an equivalent volume of aggregate.

Table 3 - Samples and mixing mass ratios used in the study

Material	Sample and Mixing ratio				
	REF	**EP 15**	**EP 30**	**EC 15**	**EC 30**
Cement	1	1	1	1	1
Sand	1.5	0.89	0.73	0.89	0.73
Perlite	-	0.16	0.32	-	-
Clay	-	-	-	1.4	2.8
Gravel	3	3	3	3	3
Water	0.48	0.48	0.48	0.48	0.48
Additive superplasticizer	2.0%	1.3%	1.2%	1.5%	2.0%

The water/cement relation is in accordance with Brazilian standard for concrete design. The class of slump was achieved with the use of a superplasticizer based on polycarboxylate, named tecflow 800, from GCP Applied Technologies brand. The proportions of EP and EC were defined as defined by Pacheco et al. [34].

Molding was performed in cylindrical (10 cm x 20 cm) and prismatic (6 cm x 6 cm x 18 cm) molds. Throughout the process, slump tests were conducted in accordance with ASTM C143 [35] and found to yield between 50 mm and 100 mm. Samples were cured both in a humidity chamber and submerged. Both curing processes were conducted under strict climate-controlled conditions with air and water temperatures of 23 ± 2ºC and relative humidity conditions of 95 ± 5%. Molding followed procedures of ASTM C470 [36].

For the prismatic samples, a 5 mm diameter CA60 steel bar segment was inserted to promote cracking in lieu of subjecting the sample to a flexural test until failure. Since the sample was 6 cm in height, the bar segment was inserted to depths of 2 cm from the base and 4 cm from the top. The prismatic samples were applied only to allow the analysis of concrete healing. Cylindrical samples were submitted to two tests: water absorption and compressive strength.

2.4 Sample Characteristics and Crack Formation

Compression strength tests were performed according to ASTM C39 [37] procedures after 7 days, 28 days and 56 days of aging with 2 samples each time.

In prismatic specimens, cracks were obtained from a flexural test according to ASTM C348-97 [38] in 7-day samples. In this case, load was applied until crack formation. The load was applied to crack formation until the value of 1.1 kN, aiming to achieve an opening of 0.3mm. After the sample's cracking, Capillary absorption tests followed the procedures of RILEM TC 116 PCD [39] after 7 days, 28 days and 56 days of aging. Test times used were of 10 min, 20 min, 30 min, 40 min, 50 min, 1 h, 2 h, 3 h, 4 h and 24 h. Also, after the crack formation, the cracks were optically evaluated with a Starret® Galileo AV 300+ Automatic model 3-D measuring apparatus. Images generated had amplifications of 62x and 164x and were post-processed in ImageJ software. Starting from calibrated values, a reference line segment of known length was created on ImageJ. This allowed the measurement of open and healed crack widths (mm) and the calculation of regenerated percent area (mm^2), as established by Wiktor and Jonkers [40]. Through the Equation 1 from the mentioned authors, it was possible to determine the healing percentage from the specimens.

$$HP(\%) = \left(\frac{w_i - w_t}{w_i}\right) \times 100 \quad (1)$$

Where:
HP (%): Healing percentage
w_i : crack initial width;
w_t : crack width in a time t.

3. RESULTS AND DISCUSSION

3.1 Mechanical strength

Table 4 presents the results of individual and potential strength resistance. As noted by Sengul et al. [24], strength decreased as EP substitution increased: a reported substitution of 20% yielded a decreased strength of around 40%. Jedidi et al. [41] also reported similar results with a 65% decrease in strength with a 30% EP substitution. Leyton-Vergara et al. [42] indicated that a maximum 40% substitution was the limit for a viable adequate concrete strength while Alazhari et al. [43] set this substitution limit at 20%.

Table 4 - Strength test results

Sample	Compressive strength (MPa)					
	7 days		28 days		56 days	
	Ind.	Potential	Ind	Potential	Ind	Potential
REF	46.5	46.5	48.5	49.0	52.6	52.6
	46.3		49		49.9	
EP 15	40.2	43.5	51.1	55.9	52.8	54.3
	43.5		55.9		54.3	
EP 30	34.8	36.6	37.6	42.3	45	45.0
	36.6		42.3		44.4	
EC 15	27.2	29.4	35.7	35.7	40.5	40.5
	29.4		34.9		34.9	
EC 30	32.1	33.6	37.2	37.2	37	37.0
	33.6		35.7		36.5	

However, for this study, 30% EP substitution resulted in a decrease in strength of approximately 13% after 28 days. Furthermore, 15% EP substitution resulted in a strength superior to the reference sample, which was rather anomalous when compared to the results of the other substitution samples. It was believed that for this case the healing agent might have filled void spaces and pores within the matrix for this sample, which would explain the increase in strength. The remaining samples agreed with the general result with the largest substitution resulting in the largest decrease in strength. It should be noted that after 56 days, the strength of the EP15 sample was very close to REF. Rashad [19] noted that EP substitution commonly had a negative impact in concrete mechanical properties but, since it was a powdered substance, might result in a gain in compaction in the final material.

In relation to compressive strength results from samples EP15 comparing with reference samples, it is noticed that they are similar. At 7 and 56 days, the potential compressive strength from EP15 samples is slightly higher than the reference. It is possible to relate those differences not only due to perlite usage, but also to the standard deviation common in cement compositions. However, the impact of perlite usage is more evident with samples named as EP30, with larger EP percentage. In all ages, the results from EP30 remain with a potential strength of up to 21% below the REF, which is higher than the applicable deviation values for concrete.

Comparing the results from EP and EC, the EP results were higher than EC samples in all ages and percentages of use, due to the lower dimension of EP. It can be pointed out that the aggregate's resistance to be used as capsules is reduced and it is not applied to enhance the composite compressive strength. EP and EC were considered as porous aggregates to be used in concrete for self-healing [6] and without the encapsulating agent works as voids inside concrete matrix. However, it is worth noting that EP presented better behavior considering the distribution of the aggregates inside concrete and also the packing analysis. Pacheco et al. [34]. Thus, due to the quality, dimension and distribution of EC, may have affected the concrete microstructure, damaging its compressive strength.

In terms of EC substitution, Shafigh et al. [44] obtained a small alteration in strength after 7 days which demonstrates a stabilizing trend. This result was observed in this study with 30% EC substitution. Despite this detrimental effect, Alghamri et al. [20] noted that light aggregates impregnated with sodium metasilicate were able to recover 80% of the resistance after cracking. This resonated well with the EP15 samples having strengths superior to REF and EC30 samples having strengths superior to EC15. Pelletier et al. [16] also observed mechanical recovery of samples containing healing agents 11% superior to reference. These results showed that, while light aggregates might impact mechanical strength negatively, post-cracking strength recovery of healing agents was superior to conventional concrete.

3.2 Capillary water absorption

Figures 5, 6 and 7 present average results of capillary water absorption after 7 days, 28 days and 56 days, respectively. The values are pointed out in Table 5. It should be noted that days were counted after the appearance of cracks from flexural tests.

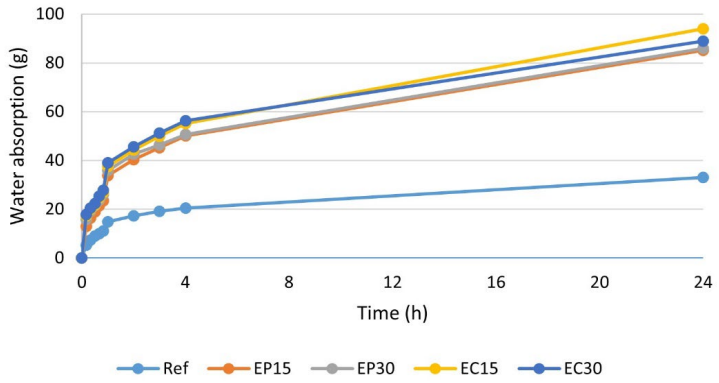

Figure 5 - Capillary water absorption after 7 days

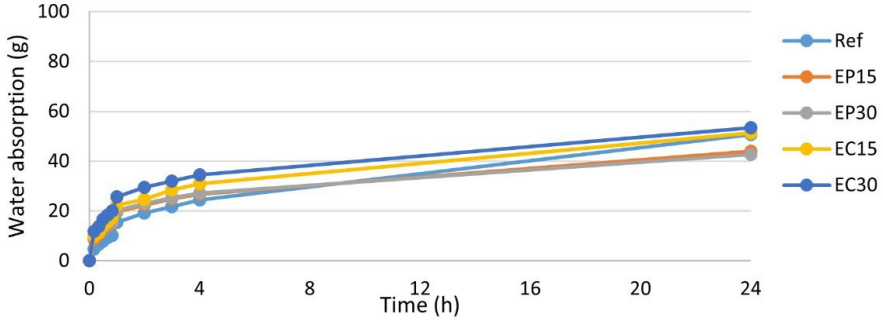

Figure 6 - Capillary water absorption after 28 days

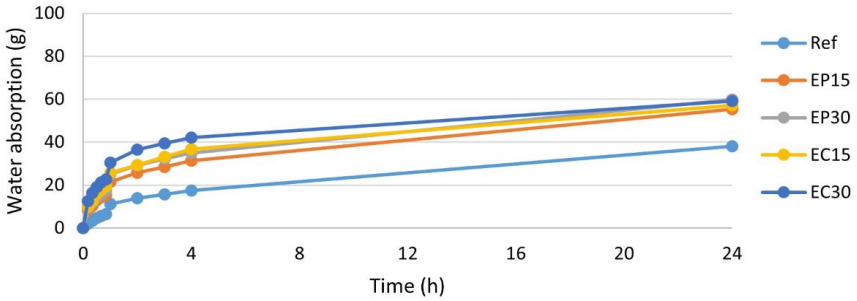

Figure 7 - Capillary water absorption after 56 days

Table 5- Water absorption (g)

T	7 days					28 days					56 days				
	R	EP		EC		R	EP		EC		R	EP		EC	
		15	30	15	30		15	30	15	30		15	30	15	30
0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
10m	5.2	13	15.8	17.2	17.8	4.7	8.7	9.8	9.8	11.8	1.9	8.4	9.4	10.1	12.6
20m	7.4	16.4	19.2	19.9	20.5	6.5	10.6	11.6	11.2	13.8	3.3	10.3	11.9	12.5	16.3
30m	9	19	21.3	21.7	22.4	7.8	12.5	13.3	13.9	16.7	4.7	12.9	14.4	15.9	19.1
40m	10	21.4	23.7	24.6	25.4	9.2	13.9	14.7	15.6	18.3	5.5	14.4	15.9	17.9	21.2
50m	11	23.5	26	26.9	27.8	10.1	14.9	15.7	17.3	20	6.4	15.5	17.5	19.4	22.6
1	14.9	33.8	35.8	37.6	39	15.3	19.5	20.2	22.3	25.6	11.2	21.5	25	25.8	30.4
2	17.2	40.3	42.6	44.3	45.6	19.1	22.4	22.9	24.7	29.4	13.9	25.8	29	29.3	36.4
3	19.1	45.3	46.4	49.9	51.2	21.7	24.8	25.2	28.5	32	15.7	28.4	32.2	33.1	39.5
4	20.5	50	50.6	55.1	56.2	24.5	26.8	27.2	30.9	34.5	17.4	31.4	34.9	36.8	42
24	33.1	85.2	85.9	93.9	88.9	50.7	44	42.7	51.4	53.4	38	55.4	59.8	57.1	59.2

As seen in Figures 5 through 7 and in Table 5, all substitution samples presented higher water absorption than the reference sample (R). This could be explained as the substitute aggregates contained void spaces which, even though the material was impregnated with healing agent, acted as water storage spaces. All substitution samples also presented the same trends with an initial absorption followed by saturation as expected [14]. As pointed out by Pacheco et al. [34] by 3D microtomography, EC mixes presented a higher void ratio when comparing with EP samples and reference mixture. In the same context, it could be noted that EP samples had less water absorption than EC samples, likely due to void spaces and packing characteristics of each mixing ratio. It was noted that from 7 to 56 days, there was a reduction of around 30% in the water absorption of the samples with healing agents. For comparison, the reference samples had an increase of around 15% in the water absorption in the same period. When comparing identical concrete samples, it should be noted that capillary water absorption naturally decreased with age. This was a result of the complete hydration of concrete samples which increased density and decreased void spaces [1], [44].

Alghamri et al. [20] obtained a reduction in absorption of 50% in samples with self-healing agents. However, it was also determined that self-healed materials were able to completely recover their permeability. This confirmed the property of sodium metasilicate to improve absorption and reduce permeability. This was likely the result of healing agent deposition.

Tan et al. [7] initially compared capillary absorption in samples without cracks and found that self-healing concrete had absorption of 0.70% while normal concrete was 1.7%. However, post-cracking, absorptions were of 0.70% and 3.54% for self-healing and normal concrete, respectively. This demonstrated that self-healing concrete maintained its initial absorption despite the crack while normal concrete increased absorption considerably. It should be noted that the use of light aggregates, even in self-healing concrete, might have increased water content inside the material.

3.3 Visual Analysis

It is worth mentioning that the results from visual analysis were obtained by superficial tests, and thus, they do not include the crack depth.

Visual analysis did not yield any healing results 7 days after cracking. However, after 28 days, samples EP15 and EP30 were observed to have healing activity and were examined with 3-D measuring equipment. Figure 8 compares the initial condition of sample EP15 (Figure 8a) with the areas to be analyzed for cracking closure percentage (Figure 8b).

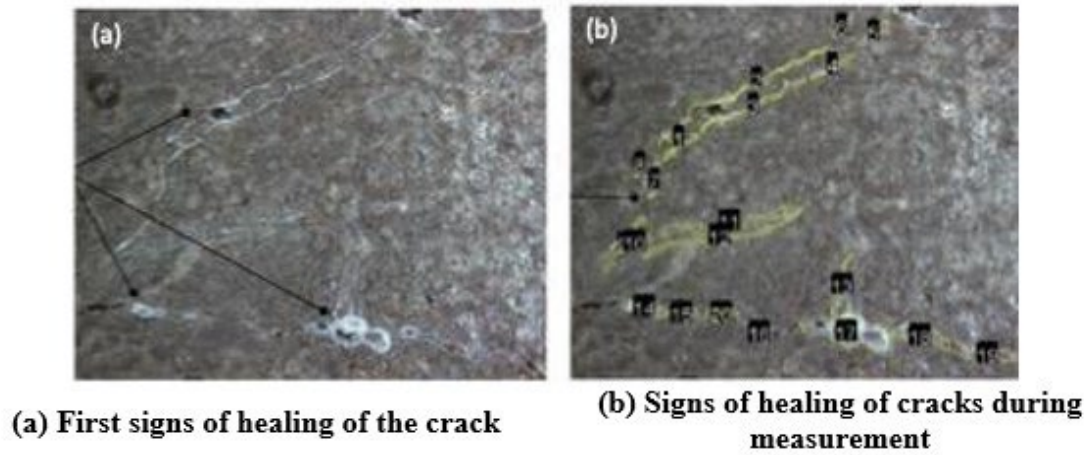

Figure 8 - Original cracking and healing activity areas to be analyzed in EP15

Close examination of cracks shown in Figure 9 further identified healing activity after 28 days in EP samples.

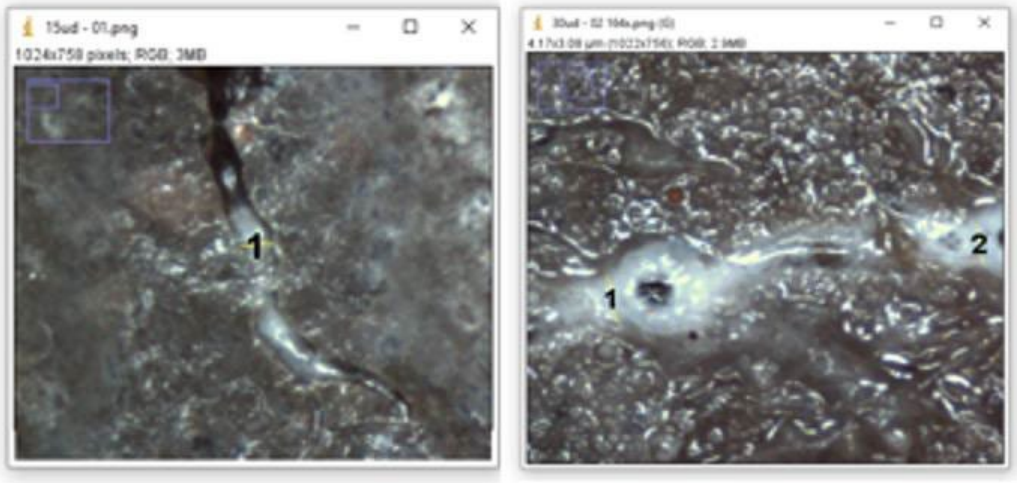

Figure 9 - Detail image of crack and healing product formation in EP15

Table 6 presents a compilation of results of initial crack opening, healing and healing percentage with respect to sample and curing conditions. The EP achieved the most healing activity at 15% substitution and that submerged curing was superior to humid curing, achieving close to total healing. In comparison, a 30% EP substitution yielded results like the other test samples. When compared to EC, EP was more efficient – a likely result of the substitute aggregate size and necessary fracture tension for the release of the healing agent. The best results of EP use in comparison to EC were aligned with the results of Zhang et al. [6]. Reductions in absorption and visual crack healing were also observed by Alazhari et al. [43] for a bacterial solution encapsulated in EP. Considering EC only, results were similar for all EC samples regardless of the degree of substitution, but it was noted that submersed EC curing yielded better results since no humid curing sample achieved more than 51% healing.

Table 6 - Cracking, healing and healing % obtained for each sample

Sample	Curing conditions	Initial crack opening (mm^2)	Healing (mm^2)		Healing percentage (%)	
			28 days	56 days	28 days	56 days
REF	Humid	0.085	0.000	0.033	0.00	38.82
REF	Submerged	0.160	0.000	0.075	0.00	46.88
EP15	Humid	0.066	0.038	0.038	57.58	57.58
EP15	Humid	0.092	0.079	0.082	85.87	89.13
EP15	Submerged	0.125	0.076	0.125	60.80	100.00
EP15	Submerged	0.141	0.118	0.134	83.69	95.04
EP30	Humid	0.109	0.106	0.109	97.25	100.00
EP30	Humid	0.106	0.000	0.088	0.00	83.02
EP30	Submerged	0.056	0.056	0.056	100.00	100.00
EP30	Submerged	0.201	0.000.	0.172	0.00	85.57
EC15	Humid	0.137	0.000	0.047	0.00	34.31
EC15	Humid	0.095	0.000	0.012	0.00	12.63
EC15	Submerged	0.036	0.025	0.032	69.44	88.89
EC15	Submerged	0.099	0.060	0.089	60.61	90.00
EC30	Humid	0.105	0.000	0.020	0.00	19.05
EC30	Humid	0.147	0.065	0.075	44.22	51.02
EC30	Submerged	0.073	0.060	0.065	82.19	89.00
EC30	Submerged	0.103	0.088	0.095	85.44	92.23

Zhang et al. [6] made similar use of image amplification equipment on 4 samples and observed crack recovery of up to 0.79 mm at 28 days. Figure 10 presents comparisons of healing progress of some samples of this study after 28 days and 56 days. As seen in Figure 10, some samples presented healing in some cracks likely due to formation of calcium silicate hydrate (C-S-H) gel. Yow and Routh [45] reported similar healing and confirmed by Alghamri et al. [20] and Sisomphon et al. [18]. Some cracks presented elevated healing potential but since the depth of the cracks was unknown, the true efficiency of healing could not be assessed.

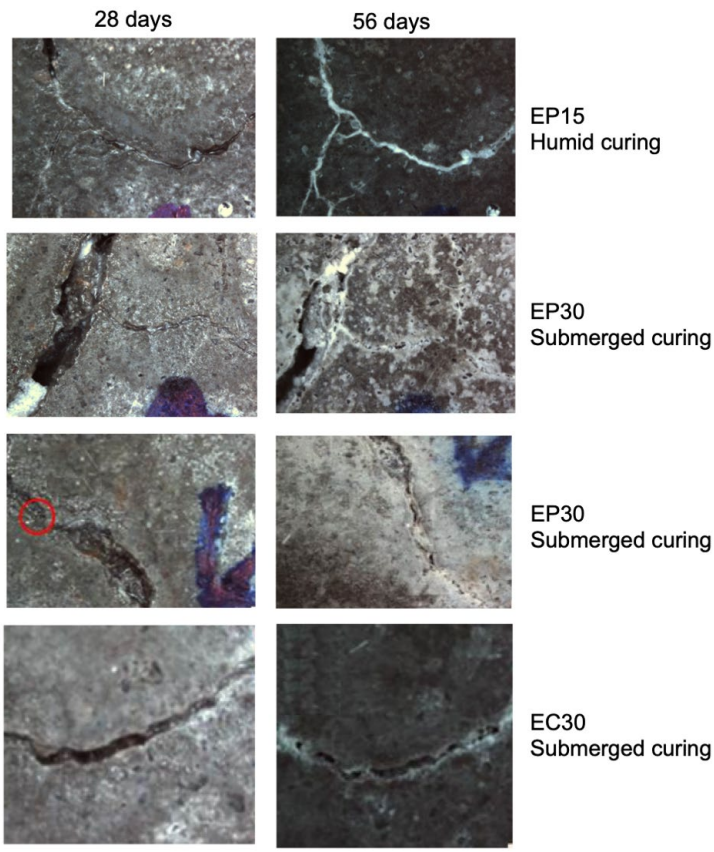

Figure 10 - Healing progress comparison at 28 days and 56 days

The images of Figure 10 suggested the possible reaction of sodium metasilicate with humidity to form products that contribute to the curing process. However, there appeared to be no linearity to the healing which, according to Van Tittelboom et al. [13], could be because healing occurred only at locations where healing material was available. In this case, locations with no visible healing activity might be the result of insufficient healing agent absorption in the substitute aggregate or a lack of rupturing to release it.

It should be noted that concrete with light aggregates tended to crack at the aggregates and not in the transition zone. This was a result of light aggregates being more susceptible to tension than compression as this relation was inversely proportional to particle size [46]. Consequently, in this study, if a crack appeared along a region of smaller particles, there was a chance that the substitute aggregate did not fracture and sodium metasilicate was not exposed to humidity. Those results of cicatrization occurrence being affected by the crack area, the surface characteristics, and the healing product availability were also pointed out by Pacheco et al. [34] and Muller et al. [47]. Overall, EP substitution achieved between 57.58% and 100% healing of the crack while EC substitution achieved between 12.63% and 90% healing. Of the 3 EP samples that achieved 100% healing, there were no observable cracks on the surface as it was completely covered in healing chemical products. As a caveat, it should be noted that optical measurements were taken at selected locations and, for most cases, did not cover the full length of the cracks present.

4. CONCLUSIONS

Substitute aggregate EP15 achieved higher mechanical strength than the reference sample for periods equal or longer than 28 days. This was likely due to crack matrix regeneration since light aggregates usually resulted in less resistance. When comparing EP and EC substitution, EP samples had superior resistances at all mixing ratios.

Capillary absorption of samples containing the healing agent was approximately 30% lower in between aging periods while the reference sample presented an increase of 15% for the same intervals. These results indicated that not only there was healing activity in the matrix but also that it had a high probability of occurring.

Optical analysis showed 100% crack area healing in 3 samples with EP substitution. The largest cracks healed were 0.056 mm for EP and 0.103 mm for EC with the latter achieving 90% healing.

Comparing both types of substitute aggregate, it was determined that EP was more efficient in absorbing sodium metasilicate so that more material is available to react with calcium hydroxide and produce C-S-H vital to concrete and its properties.

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